

# PATENT SPECIFICATION

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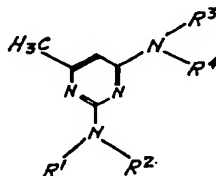
## (54) 2,4-DIAMINOPYRIMIDINE DERIVATIVES AND THEIR USE AS PESTICIDES

(71) We, WACKER—CHEMIE G.M.B.H., a body corporate organised according to the laws of the Federal Republic of Germany, of 8 München 22, Prinzregentenstrasse 22, Federal Republic of Germany, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to 2,4-diamino-6-methyl-pyrimidines, which are useful as pesticides.

Many pesticidally active, especially herbicidally active, compounds are known, but most of these are active either against broad-leaved plants or against grasses, but not both. The present invention now provides 2,4-diamino-6-methyl-pyrimidines that are, in most cases, active against both broad-leaved plants and grasses. They also exhibit a fungicidal action as well as a herbicidal one.

The 2,4-diamino-6-methyl-pyrimidines of the invention are of the general formula



in which each of R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup>, which may be identical or different, denotes a hydrogen atom or an alkyl group (straight or branched chain) having up to 8 carbon atoms, with the proviso that at least one of the said symbols denotes a hydrogen atom, that at least one of the said symbols denotes an alkyl group, and that the total number or carbon atoms in an alkyl groups denoted by any of the said symbols is from 6 to 12, preferably from 7 to 10. The compounds of the invention can also be used in the form of their salts, for example their hydrochlorides or hydrobromides.

An optimum fungicidal action is obtained with compounds in which the total number of carbon atoms in the alkyl groups R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> is 10, with each amino nitrogen atom carrying at least one alkyl group (that is to say that at least R<sup>1</sup> and R<sup>3</sup> should denote alkyl groups).

A good herbicidal action is generally obtained with those compounds in which only one of the symbols R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> denotes a hydrogen atom. A particularly good herbicidal activity is shown by those compounds in which R<sup>1</sup> denotes a hydrogen atom, R<sup>2</sup> denotes a pentyl or hexyl group (straight or branched), R<sup>3</sup> denotes a methyl group, and R<sup>4</sup> denotes a methyl or ethyl group. The total number of carbon atoms in the alkyl groups R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> denotes a methyl or ethyl group. The total number of carbon atoms in the alkyl groups R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> is generally 8.

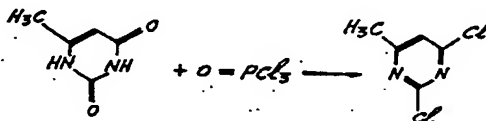
The following are specific examples of pyrimidines according to the invention:

2,4-bis-n-propylamino-6-methylpyrimidine,  
2,4-bis-isopropylamino-6-methylpyrimidine,  
2,4-bis-n-butylamino-6-methylpyrimidine,  
2,4-bis-isobutylamino-6-methylpyrimidine,  
2,4-bis-1',2'-dimethylpropylamino-6-methylpyrimidine,

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2,4-bis-1',2'-dimethylbutylamino-6-methylpyrimidine,  
 2-amino-4-ethylhexylamino-6-methylpyrimidine and its isomer,  
 2-dimethylamino-4-n-hexylamino-6-methylpyrimidine and its isomer,  
 2-dimethylamino-4-1',3'-dimethylbutylamino-6-methylpyrimidine and its isomer,  
 2-ethylamino-4-1',3'-dimethylbutylamino-6-methylpyrimidine and its isomer,  
 2-ethylamino-4-n-hexylamino-6-methylpyrimidine and its isomer,  
 2-n-propylamino-4-n-butylamino-6-methylpyrimidine and its isomer,  
 2-isopropylamino-4-n-butylamino-6-methylpyrimidine and its isomer,  
 2-isopropylamino-4-isobutylamino-6-methylpyrimidine and its isomer,  
 2-n-propylamino-4-isobutylamino-6-methylpyrimidine and its isomer,  
 2-n-butylamino-4-isobutylamino-6-methylpyrimidine and its isomer,  
 2-n-butylamino-4-n-pentylamino-6-methylpyrimidine and its isomer,  
 2-n-butylamino-4-n-hexylamino-6-methylpyrimidine and its isomer,  
 2-iso-butylamino-4-1',3'-dimethylbutylamino-6-methylpyrimidine and its isomer,  
 2-n-butylamino-4-1',3'-dimethylbutylamino-6-methylpyrimidine and its isomer,  
 2-isobutylamino-4-n-pentylamino-6-methylpyrimidine and its isomer,  
 In the above list the reference to isomers means the compounds in which the substituents in the 2-position and 4-position respectively are interchanged.

The compounds of the invention can be manufactured by the amination of 2,4-dichloro-6-methylpyrimidine, which is obtainable from 6-methyluracil. 6-methyluracil may be reacted with phosphorus oxychloride, which can also serve as the solvent, to give 2,4-dichloro-6-methylpyrimidine in known manner:



The 2,4-dichloro-6-methylpyrimidine is then aminated with an amine of the general



and with an amine of the general formula



in either order. Amination of the first chlorine atom takes place very exothermically and will take place even at low temperatures, although for economic reasons it is generally not carried out below 0° C. Amination of the second chlorine atom can be achieved only at temperatures above 90° C., preferably above 120° C. Suitable solvents for the amination are chlorinated hydrocarbons (e.g. chlorobenzene and dichlorobenzene), hydrocarbons (e.g. toluene and o-xylene), phenol, water, alcohols (e.g. methanol, ethanol, propanol, butanol, hexanol, heptanol, octanol, glycol, glycerol, propanediol, and butanediol), and mixtures of alcohol and water. A solvent is often not necessary for the second amination step. This process can be carried out in two stages, with amination of the first chlorine atom being carried out at a temperature below 90° C. and that of the second chloride atom being carried out at a temperature above 90° C., and it is therefore particularly suitable for the manufacture of these compounds of the invention having a substituent in the 2-position that is different from that in the 4-position. The process can of course be used for the manufacture of compounds having identical substituents in these two positions, the above two amines being identical, in which case there is no need to aminate the two chlorine atoms separately and the entire reaction may therefore be carried out at a temperature above 90° C.

Another method of making compounds of the invention in which the substituents in the 2-position and in the 4-position are identical is by reacting 6-methyluracil, with a phosphoric triamide of the general formula



or with a phosphorous triamide of the general formula



to give a 2,4-bis-alkylamino-6-methylpyrimidine in which  $R^1$  and  $R^2$  both denote hydrogen atoms and  $R^3$  and  $R^4$  denote identical alkyl groups. This reaction is carried out at a temperature of from 130 to 280° C., and a reaction time of from 0.5 to 4 hours is generally required. Amine hydrochlorides are produced during the manufacture of the triamides and these can usefully serve as catalysts for the amination of the 6-methyluracil.

The 2,4-diamino-6-methylpyrimidines of the invention can be separated from the various by-products, such as phosphoric amides, phosphoric acid, and amine hydrochlorides, by the use of various organic solvents, for example, chlorinated hydrocarbons (e.g. carbon tetrachloride, chloroform, and methylene chloride), hydrocarbons (e.g. benzene and toluene), and ethers (e.g. diethyl ether, dibutyl ether, and tetrahydrofuran).

The compounds of the invention can be used *per se* as pesticides, but they are more usually formulated into pesticidal preparations containing from 0.005 to 80% by weight of the 2,4-diamino-6-methylpyrimidines or salts thereof. The manner of formulation will depend on the desired physical form of the pesticidal preparation, which will in turn depend on the desired method of application of the preparation. The preparations can, for example, be in the form of emulsion concentrates, wettable powders, or dusting agents. These preparations may then be diluted as required and applied, for example, by dusting, scattering, spraying, or atomising, to crops or a crop area.

A suitable formulation for an emulsion concentrate is, for example, from 10 to 50% by weight of the 2,4-diamino-6-methylpyrimidine or salt thereof, from 35 to 80% by weight of an organic solvent (e.g. benzene, toluene, xylene, cyclohexanone, isopropanol, butanol, glycol ethers, ethoxymethanol, or butoxymethanol), and from 5 to 15% by weight of a dispersing auxiliary (e.g. a sodium alkylbenzenesulphonate, calcium dodecylbenzenesulphonate, an alkyl polyglycol ether, an alkylphenol/ethylene oxide condensation product, or a sodium alkylnaphthalenesulphonate). The concentrate can be prepared by intimately mixing the components in a stirring vessel.

Wettable powders may, for example, be formulated from 30 to 80% by weight of the 2,4-diamino-6-methylpyrimidine or salt thereof, from 1 to 10% by weight of a dispersing auxiliary (e.g. those mentioned above), and from 10 to 60% by weight of an inert filler (e.g. kaolin, montmorillonite, china clay, magnesium carbonate, calcium carbonate, kieselguhr, or highly disperse silica).

Dusting agents generally contain from 5 to 25% by weight of the 2,4-diamino-6-methylpyrimidine or salt thereof, the remainder consisting of inert filler (e.g. those mentioned above).

The pulverulent preparations, such as wettable powders and dusting agents, are conveniently prepared by intimately mixing the various components together, and then grinding the mixture by means of a hammer mill or other grinding device to a particle size generally less than 20  $\mu$ m. The ground mixture is again mixed and then sieved.

The pesticidal preparations according to the invention can contain other pesticidally active compounds in addition to the 2,4-diamino-6-methylpyrimidines. In this way an increased pesticidal action or a better toleration by crops can sometimes be achieved. Examples of such other pesticidally active compounds are ureas, aryloxy-fatty acids, triazines, carbamates, thiocarbamates, dinitroalkylanilines, acylanilides, and dinitrophenols.

The following Examples illustrate various aspects of the invention. All percentages are by weight.

#### *Manufacture of compounds.*

The compounds listed in Table 1 were prepared according to the following procedures, and their structures were analysed by NMR spectroscopy.

**Procedure 1:** Preparation of 2-(or 4-)alkylamino-4-(or 2-)chloro-6-methylpyrimidine.

0.5 mole of 2,5-dichloro-6-methylpyrimidine was partially dissolved in about 500 ml of ethanol or a water/ethanol mixture, and 1 mole of the desired amine was added dropwise to the suspension over the course of 1 to 3 hours at 0 to 20° C. The reaction was in most cases slightly exothermic and was kept within the given temperature range by cooling. The resulting solution was left to stand at room temperature for several days and was then concentrated in a rotary evaporator. The residue was shaken with water and chloroform and the aqueous and organic phases were separated.

*Procedure 2:* Preparation of 2,4-bis-alkylamino-6-methylpyrimidines with different alkylamino groups.

0.5 mole of (A) 4-alkylamino-2-chloro-6-methylpyrimidine or (B) 2-alkylamino-4-chloro-6-methylpyrimidine, prepared according to procedure 1, was heated to about 150° C. and 1 mole of the desired amine was added dropwise very slowly, after which the mixture was maintained at from 120 to 150° C. for 4 to 6 hours. The resulting mixture was cooled, and shaken with water and chloroform. The layers were separated, and the organic phase was dried with sodium sulphate or potassium carbonate. The drying agent was then filtered off. The filtrate was stripped of solvent in a rotary evaporator, and the residue was distilled under a high vacuum.

*Procedure 3:* Preparation of 2,4-bis-alkylamino-6-methylpyrimidines with identical alkylamino groups.

0.8 mole of the desired amine was added dropwise to 0.2 mole of 2,4-dichloro-6-methylpyrimidine. The reaction was initially very exothermic, until about 40% of the amine had been used, but the mixture was not cooled unless its temperature exceeded 150° C. The remaining 60% of the amine was added dropwise very slowly at 130 to 160° C., after which the mixture was maintained at from 120 to 150° C. for from 4 to 6 hours. The mixture was cooled, and then shaken with water and chloroform. The organic phase was separated from the aqueous phase and dried with potassium carbonate and sodium sulphate, which was subsequently filtered off. The solvent was removed from the filtrate in a rotary evaporator, and the residue was distilled in a high vacuum.

*Procedure 4:* Preparation of 2,4-bis-n-butylamino-6-methylpyrimidine.

0.5 mole of phosphoric tri-n-butylamide and 0.25 mole of 6-methyluracil were mixed and heated at 200° C. for 0.5 hour. The mixture was cooled. Chloroform was then added and the mixture was washed with sodium carbonate solution. The chloroform layer was separated, and the solvent was stripped off. The residue was distilled under a pressure of 0.5 to 0.3 mm Hg, to give a product boiling at 134—157° C.

TABLE 1

Example	R <sup>1</sup> R <sup>2</sup> R <sup>3</sup> R <sup>4</sup>				Procedure	Yield %	Analysis					
	calculated: C H N	found: C H N										
1	H	n-C <sub>3</sub> H <sub>7</sub>	H	n-C <sub>3</sub> H <sub>7</sub>	3	84	63.4	9.7	26.9	63.8	10.0	27.0
2	H	iso-C <sub>3</sub> H <sub>7</sub>	H	iso-C <sub>3</sub> H <sub>7</sub>	3	78	63.4	9.7	26.9	63.7	9.7	25.2
3	H	n-C <sub>4</sub> H <sub>9</sub>	H	n-C <sub>4</sub> H <sub>9</sub>	3/4	61/40	66.1	10.2	23.7	65.2	9.8	24.3
4	H	-CHCH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	H	-CHCH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	3	52	-	-	-	-	-	-
5	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	3	77	-	-	-	-	-	-
6	H	n-C <sub>3</sub> H <sub>7</sub>	H	n-C <sub>4</sub> H <sub>9</sub>	2 B	77	64.8	10.0	25.2	64.2	10.2	24.9
7	H	n-C <sub>4</sub> H <sub>9</sub>	H	n-C <sub>3</sub> H <sub>7</sub>	2 A	86	64.8	10.0	25.2	64.0	10.1	25.0
8	H	C <sub>2</sub> H <sub>5</sub>	H	n-C <sub>6</sub> H <sub>13</sub>	2 B	65	66.1	10.2	23.7	65.3	10.5	22.5
9	H	n-C <sub>6</sub> H <sub>13</sub>	H	C <sub>2</sub> H <sub>5</sub>	2 A	87	66.1	10.2	23.7	66.2	10.3	23.1
10	H	n-C <sub>4</sub> H <sub>9</sub>	H	n-C <sub>4</sub> H <sub>9</sub>	2 A	81	68.1	10.7	21.2	67.7	10.9	21.0
11	H	n-C <sub>6</sub> H <sub>13</sub>	H	n-C <sub>4</sub> H <sub>9</sub>	2 B	80	68.1	10.7	21.2	68.8	10.4	20.3
12	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	H	n-C <sub>4</sub> H <sub>9</sub>	2 A	93	68.1	10.7	21.2	67.2	10.0	20.6
13	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	H	-CH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>	2 B	84	-	-	-	-	-	-

TABLE 1 (Continued)

Example	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	Procedure	Yield %	Analysis		
							calculated:	found:	
							C	H	N
14	H	n-C <sub>6</sub> H <sub>13</sub>	CH <sub>3</sub>	CH <sub>3</sub>	2 A	86	66.1	10.2	23.7
15	CH <sub>3</sub>	CH <sub>3</sub>	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	2 B	83	66.1	10.2	23.7
16	H	-CHCH <sub>2</sub> CH(CH <sub>3</sub> ) <sub>2</sub>   CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	2 A	74	66.1	10.2	23.7
							65.6	10.3	23.1
							66.3	10.1	22.7
							65.1	9.7	19.0

*Formulation of pesticides.*

The following formulations 1—3 are examples of the manner in which the compounds of the invention can be formulated into pesticidal preparations.

5

*Formulation 1: Emulsion concentrate.*

Active compound 10 to 50%  
Cyclohexanone 20 to 60%  
Xylene 5 to 20%  
Emulsifier 1HF 5 to 15%

10 (Emulsifier 1HF is a mixture of a sodium alkylbenzenesulphonate, an alkyl polyglycol ether, and solvent. It is obtainable from Chemische Werke Hüls, Federal Republic of Germany.)

10

*Formulation 2: Wettable powder.*

Active compound 30 to 80%  
Sodium dioctyl sulphosuccinate 2 to 3%  
Sodium ligninsulphonate 0 to 4%  
Highly disperse silica 0 to 3%  
Kaolin 10 to 60%

15

*Formulation 3: Dusting agent.*

Active compound 5 to 25%  
Highly disperse silica 0 to 1%  
Calcium carbonate 70 to 95%

20

**Pesticidal properties.**

The pesticidal properties of the compounds according to the invention are demonstrated by the following two tests.

**Test 1: Herbicidal properties.**

- 5 The following plants were sown in plant dishes in a greenhouse: *Zea Mays* (maize), *Hordeum* (barley), *Digitaria* (millet), *Sinapis* (mustard), *Centaurea* (cornflower), *Galium aparine* (cleavers), *Beta* (sugar beet). 14 days after emergence, the plants were sprayed with 6 kg/ha of emulsion concentrate: 5

- 10 active compound 20%  
cyclohexanone 50%  
xylene 20%  
emulsifier 1HF 10%  
(Emulsifier 1HF is described in Formulation 1.) 10

- 15 The results, in terms of the number of plants destroyed, were assessed after 28 days and are shown in the following Table 2, for each of the compounds prepared according to Examples 1 to 16. In the Table, 0 indicates no activity, i.e. no plants destroyed, and 10 indicates 100% of plants destroyed. 15

The herbicidal action of compounds 14 to 16 is particularly good. These two compounds also show a good action against wild oats, and a good defoliant action.

TABLE 2

No.	Barley	Millet	Maize	Mustard	Sugar Beet	Cleavers	Cornflower
1	3	3	0	8	7	8	8
2	3	7	7	8	5	6	6
3	6	8	9	10	10	9	10
4	4	3	4	8	4	3	9
5	4	5	5	5	1	2	5
6	1	1	1	6	6	8	6
7	1	3	2	5	5	6	4
8	5	4	5	7	9	7	7
9	5	4	5	7	6	6	8
10	4	7	3	6	4	6	7
11	5	6	5	5	6	7	8
12	5	7	6	6	5	6	7
13	2	5	6	7	1	3	3
14	7	5	8	7	8	9	6
15	2	5	6	4	3	6	7
16	6	8	8	9	10	7	10

*Test 2: fungicidal properties.*

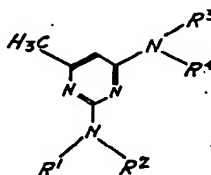
The compounds produced according to Examples 2, 3 and 11 were found, at a concentration of 0.1%, to show a good activity against leaf spot of celery (*Septoria*) and against bean rust (*Uromyces fabae*).

5

WHAT WE CLAIM IS:—

1. Compound of the general formula

5



10

in which each of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$ , which may be identical or different, denotes a hydrogen atom or an alkyl group having up to 8 carbon atoms, with the provisos that at least one of the said symbols denotes a hydrogen atom, that at least one of the said symbols denotes an alkyl group, and that the total number of carbon atoms in all alkyl groups denoted by any of the said symbols is from 6 to 12.

10

15

2. Compound as claimed in claim 1, in which the total number of carbon atoms in all alkyl groups denoted by any of  $R^1$ ,  $R^2$ ,  $R^3$  and  $R^4$  is from 7 to 10.

3. Compound as claimed in claim 1, in which at least  $R^1$  and  $R^3$  denote alkyl groups and in which the total number of carbon atoms in all alkyl groups denoted by any of  $R^1$ ,  $R^2$ ,  $R^3$ , and  $R^4$  is 10.

15

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4. Compound as claimed in claim 1, in which only one of  $R^1$ ,  $R^2$ ,  $R^3$ , and  $R^4$  denotes a hydrogen atom.

5. Compound as claimed in claim 1, in which  $R^1$  denotes a hydrogen atom,  $R^2$  denotes a pentyl or hexyl group,  $R^3$  denotes a methyl group, and  $R^4$  denotes a methyl or ethyl group.

20

25

6. Compound as claimed in claim 5, in which the total number of carbon atoms in the alkyl groups denoted by  $R^2$ ,  $R^3$ , and  $R^4$  is 8.

7. Compound as claimed in claim 1, and specifically listed herein.

25

8. Compound as claimed in claim 1 and listed in Table 1 herein.

9. Salt of a compound as claimed in any one of claims 1 to 8.

30

10. Hydrobromide or hydrochloride salt of a compound as claimed in any one of claims 1 to 8.

11. Process for the manufacture of a compound as claimed in claim 1, which comprises reacting 2,4-dichloro-6-methylpyrimidine with an amine of the general formula

30



and with an amine of the general formula

35



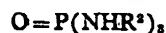
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in either order, the reaction with the second-reacted amine being arrived at at a temperature above 90° C.

40

12. Process for the manufacture of a compound as claimed in claim 1 in which  $R^1$  and  $R^3$  both denote hydrogen atoms and  $R^2$  and  $R^4$  denote identical alkyl groups, which comprises reacting 6-methyluracil with a phosphoric triamide of the general formula

40



or with a phosphorous triamide of the general formula



45

at a temperature of from 130 to 280° C.

45

13. Process as claimed in claim 11, carried out substantially as described in Procedures 1 and 2 or Procedure 3 herein.



14. Process as claimed in claim 12, carried out substantially as described in Procedure 4 herein.
15. Compound as claimed in claim 1, whenever prepared by a process as claimed in any one of claims 11 to 14 herein.
- 5 16. Pesticidal preparation containing from 0.005 to 80% by weight of a compound or salt as claimed in any one of claims 1 to 10 and 15. 5
17. Preparation as claimed in claim 16 in the form of an emulsion concentrate, wettable powder, or dusting agent.
- 10 18. Pesticidal preparation in the form of an emulsion concentrate comprising from 10 to 50% by weight of a compound or salt as claimed in any one of claims 1 to 10 and 15, from 35 to 80% by weight of an organic solvent, and from 5 to 15% by weight of a dispersing auxiliary. 10
- 15 19. Pesticidal preparation in the form of a wettable powder comprising from 30 to 80% by weight of a compound or salt as claimed in any one of claims 1 to 10 and 15, from 1 to 10% by weight of a dispersing auxiliary, and from 10 to 60% by weight of an inert filler. 15
- 20 20. Pesticidal preparation in the form of a dusting agent comprising from 5 to 25% by weight of a compound or salt as claimed in any one of claims 1 to 10 and 15, and from 75 to 95% by weight of an inert filler.
- 20 21. Pesticidal preparation as claimed in claim 16, formulated according to any one of Formulations 1 to 3 herein. 20
- 25 22. Method of combating pests, which comprises applying to crops or a crop area a pesticidal preparation as claimed in any one of claims 18 to 21.
- 25 23. Method of combating pests, which comprises applying to crops or a crop area a compound or salt as claimed in any one of claims 1 to 10 and 15. 25

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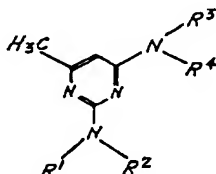
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32Y 456 45X 45Y 618 630 670 774 776 791 79Y NG



(71) We, WACKER-CHEMIE G.M.B.H., a body corporate organised according to the laws of the Federal Republic of Germany, of 8 München 22, Prinzregentenstrasse 22, Federal Republic of Germany, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

Many pesticidally active, especially herbicidally active, compounds are known, but most of these are active either against broad-leaved plants or against grasses, but not both. The present invention now provides 2,4-diamino-6-methyl-pyrimidines that are, in most cases, active against both broad-leaved plants and grasses. They also exhibit a fungicidal action as well as a herbicidal one.

The 2,4-diamino-6-methyl-pyrimidines of the invention are of the general formula



An optimum fungicidal action is obtained with compounds in which the total number of carbon atoms in the molecule is 10-12 (the nitrogen atom carrying at least one hydrogen atom should denote alkyl groups).

**SPECIFICATION No. 1,488,093**

The following are specific

2,4-bis-n-propylamino-6-meth

2,4-bis-isopropylamino-6-meth

2,4-bis-n-butylamino-6-methyl

2,4-bis-isobutylamino-6-methy

2,4-bis-1',2'-dimethylpropylam

Page 1, line 8, *for* active read active  
 Page 1, line 19, *after* in delete an insert all  
 Page 1, line 30, *after* group. (second occurrence) delete The total number of carbon atoms in  
 Page 1, line 31, delete the alkyl groups R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> denotes a methyl or ethyl group.  
 Page 5, Table 1, Example 10, Heading R<sup>4</sup>, delete n-C<sub>4</sub>H<sub>9</sub>, insert n-C<sub>6</sub>H<sub>13</sub>  
 Page 8, line 36, *after* being delete arrived at insert carried out

THE PATENT OFFICE  
3rd January, 1978